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METAL POLLUTION ASSESSMENT THROUGH AQUATIC SEDIMENTS



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CONTENTS

	Page No.
LIST OF TABLES	i
LIST OF FIGURES	ii
ABSTRACT	iii
1.0 INTRODUCTION	1
1.1 General	1
1.2 Soluble/Solid Equilibrium	2
1.3 Scope of the Study	4
2.0 MEDIA OF POLLUTION ASSESSMENT	5
3.0 METHODOLOGY	10
3.1 Sampling	10
3.2 Storage	13
3.3 Mechanical Analysis	13
3.4 Mineralogical Analysis	14
3.5 Sediment Digestion	14
4.0 REVIEW	17
4.1 Geochemical Reconnaissance of Aquatic Sediments	17
4.2 Natural Metal Content	19
4.3 Metal Pollution in Lake Sediments	20
4.4 Metal Pollution in River Sediments	23
5.0 CONCLUSION	36
REFERENCES	37

LIST OF TABLES

S.No.	Title	Page No.
1.	Percentages in particulate associated metals of total metal discharge (solid+aqueous phases) in polluted rivers in the USA and Europe	5
2.	Cadmium concentrations in sediments of rivers and streams in Europe	26
3.	Mercury concentrations in sediments of rivers and streams in North America	26

LIST OF FIGURES

S.No.	Title	Page No.
1.	Concentration of mercury in surface and bottom waters and in sediments from lake Ontario	3
2.	Distribution of arsenic in stream sediments in England	7
3.	Schematic sequence for analytical procedures in metal pollution assessment from sediments	11

ABSTRACT

The introduction of metallic pollutants into a river, whether it is natural (erosion of its drainage basin) or artificial (anthropogenic), can occur in dissolved or particulate form. Depending on physico-chemical conditions, the pollutants in dissolved form can later precipitate as hydroxides, carbonates or other minerals. They can also be adsorbed by the iron or manganese oxides and hydroxides or coprecipitate with these, or form dissolved organic or organic complexes. In particulate form, associated with suspended matter, the pollutant can, under favorable hydraulic conditions, be deposited on the bottom sediments. Therefore, the study of metallic pollutant concentrations in sediments can be conveniently used to investigate possible pollution; the contents are generally well above detection limits which is not always the case in water. Furthermore, the analysis of sediments eliminates the problem of erratic fluctuations which are often observed in water.

The sediment existing at the bottom of the water body play a major role in the pollution schemes of the river and lake systems. They reflect the current quality of the water system and can be used to detect the presence of contamination that do not remain soluble after discharge into surface water. Moreover, sediments act as carriers and possible sources of pollution because heavy metals are not permanently fixed by them and can be released back to the water column by changes in environmental conditions. Therefore, the analysis of heavy metals in sediments permits us to detect pollution that could escape water analysis, and also provides information about the critical sites of the water system under consideration.

In the present report, an attempt has been made to compile and evaluate the present state of affairs with regard to metal pollution assessment through aquatic sediments in rivers and lakes.

1.0 INTRODUCTION

1.1 General

Heavy metals today have a great ecological significance due to their toxicity and accumulative behavior (Purves, 1985). These elements, contrary to most pollutants, are not biodegradable and undergo a global eco-biological cycle (Nurnberg, 1984) in which natural waters are the main pathways. The determination of the concentration levels of heavy metals in these waters, as well as the elucidation of the chemical forms in which they appear is a prime target in environmental research today (Nurnberg, 1983; Leppard, 1983; Patterson and Passino, 1987), because of the close relation between toxicity and speciation (Smies, 1983).

The introduction of metal contaminants into the aquatic system has various sources including smelting processes and fuel combustion via atmospheric fallout, pollution from leaks, effluents and dumping activities, land application of sewage materials and leaching of garbage etc. On the other hand, polluted water bodies lead over many pathways to metal contamination of terrestrial ecosystems, for example, by way of irrigation, dredging activities and biota flux. Depending on physico-chemical conditions, the pollutants in dissolved form can later precipitate as hydroxides, carbonates or other minerals. They can also be adsorbed by the iron or manganese oxides and hydroxides or coprecipitate with these, or form dissolved organic or organic complexes (Salomons and Forstner, 1984; Drever, 1988; Buffle, 1990). Therefore, the analysis of heavy metals in sediments permits us to detect pollution that could escape water analysis, and also provides information about the critical sites of the water system under consideration (Forstner and Solomons, 1980).

The tracing of pollution sources by means of water analysis frequently gives rise to difficulties which may usually be ascribed to sampling procedures and the physical chemical condition pertaining to the investigated species rather than to

the accuracy and precision of analytic techniques. Determination of heavy metal concentrations in surface water samples collected from rivers at short intervals reveals fluctuations of several orders of magnitude. These may be partially explained by variations resulting from water discharges and the predominance of certain source areas leading to irregular effluent emissions. Exchange processes between interstitial and surface water effect fluctuations along the river course. These processes are influenced by pH and Eh conditions, the type and quality of complexing agents, their biologic activity and by salinity and temperature. Therefore, in addition to having limited experimental data available, the extremely complex system of influencing factors generally makes it impossible to arrive at any definite conclusion regarding the source of pollution.

1.2 Soluble/Solid Equilibrium

A special difficulty arises from contaminants which are not readily soluble but become rapidly fixed to particulate matter in the receiving water body. This applies, in particular, to the various heavy metals like chromium, mercury, lead etc. Even close to the point to the input, the metal content in water decreases to its normal level making detection difficult, except by means of a very close knit system of water sampling. Fig. 1 shows the distribution of mercury in surface and in bottom water (Chau and Saitoh, 1973) as well as in sediments (Thomas, 1972) of the lake Ontario.

It is clearly evident from the figure that no relationship exists between the mercury distribution in surface and bottom water; it is also impossible to pinpoint a specific pollution centre from the lateral distribution of the water data. In contrast, metal contents in the sediments reveal quite different distribution patterns, having a distinct grouping of elevated mercury levels near the southern bank of the lake, especially close to the mouth of the Niagara river. Detailed investigations carried out by Fitchko and Hutchinson (1975) have in fact shown that the dispersion pathways point to the Niagara river as the prime source of mercury input of lake Ontario. The

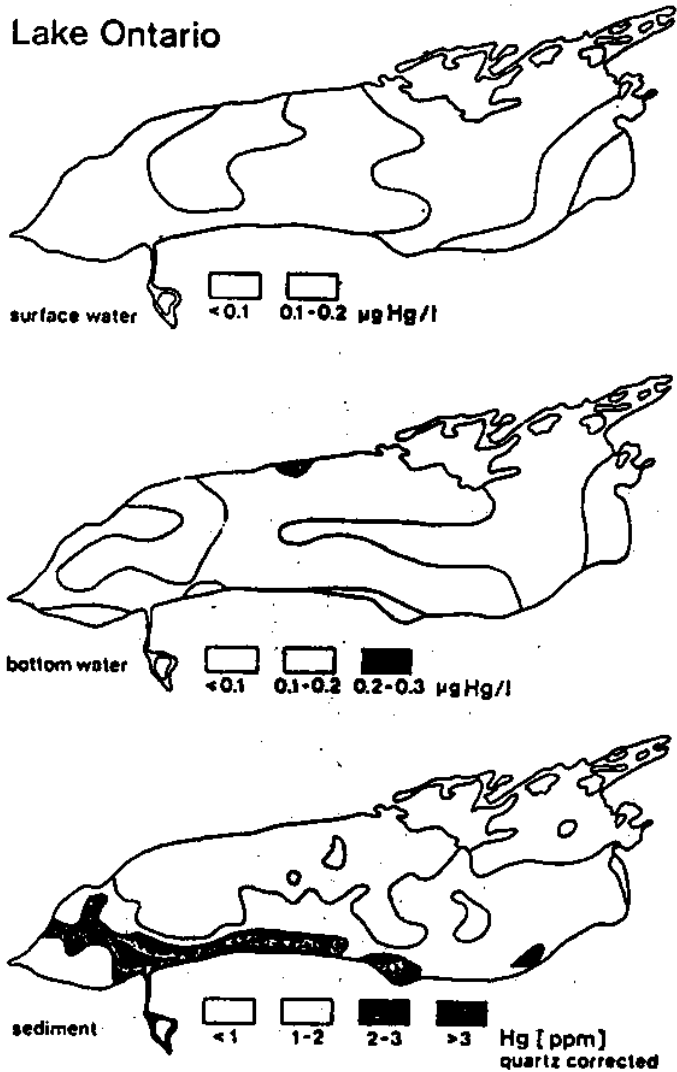


Fig. 1. Concentration of mercury in surface and bottom waters and in sediments from lake Ontario

eastward extension of the Niagara mercury plume is reported due to coastal currents (Simons, 1972), whereas the enrichment of mercury at the mouth of the Genesee and Oswego rivers appears to result from the contamination within the catchment areas.

The study of metallic pollutants in sediments can be conveniently used to investigate possible pollution; the contents are generally well above detection limits which is not always the case in water. Furthermore, the analysis of sediments eliminates the problem of erratic fluctuations which are often observed in water (Grimshaw et al., 1976; Salomons and Forstner, 1984; Erel et al., 1991; Giger et al., 1992; Bourg and Bertin, 1995).

The sediment existing at the bottom of the water body play a major role in the pollution schemes of the river and lake systems (Forstner, 1985). The establishment of metal levels in sediments can be used in detecting sources of pollution in aquatic systems. Although sediment analysis do not represent the extent of intoxication, they can be employed on a semi quantitative basis in comparative studies to trace the sources of pollution, such as surreptitious discharges by factories. Under favorable conditions, pollution sources can even be detected long after input has taken place. Furthermore, it is possible to determine their developmental of pollution intensity from dated sediment cores provided they contain fine grained depositions, in which the sorbed, precipitated, organically bonded metal concentrations are accumulated. Finally, an investigation of sediment particles is likely to be useful from an analytical point of view since trace element content in particulate matter is 1,000 to 100,000 times higher than the corresponding metal content in the associated aqueous phase.

1.3 Scope of the Study

The present report is an attempt by the author to compile and evaluate the present state of affairs with regard to metal pollution assessment through aquatic sediments in rivers and lakes. The study represents an interesting concept to study the metal pollution assessment through aquatic sediments.

Various media are analyzed to assess, monitor and control metal pollution. The most obvious medium is surface water. However, it has been established that for a given sampling station monitored over a long period of time the values of the collected samples tend to vary by several orders of magnitude, although the samples may have been collected at short-time intervals. Such fluctuations are attributable to a large number of variables, such as daily and seasonal variations in water flow, surreptitious local discharges of effluent, changing pH and redox conditions, the input of treated secondary sewage, detergent levels, salinity and temperature.

Pollutant concentrations in particulate matter often provide a more stable and convenient means of obtaining an indication of the state of associated waters. In a number of polluted rivers of the United States and Central Europe (De Groot et al., 1973b; Heinrichs, 1975), the amount of heavy metals in water and particulates were determined for a certain specified period of time. From the ratios of heavy metals in water to those adsorbed to particulates, a sequence of mobility was deduced, which is characterized by the examples in Table 1.

Table 1. Percentages in particulate associated metals of total metal discharge (solid + aqueous phases) in polluted rivers in the USA and Europe

Metals	U.S.Rivers	F.R.G.Rivers	Rhine(Neth)
Sodium	-	0.5%	-
Calcium	-	2.5%	-
Strontium	21%	-	-
Boron	30%	-	-
Cadmium	-	30%	45%
Zinc	40%	45%	37%
Copper	63%	55%	64%
Mercury	-	59%	56%
Chromium	76%	72%	70%
Lead	84%	79%	73%
Aluminium	98%	98%	-
Iron	98%	98%	-

The alkali and alkaline earth metals are predominantly present in a dissolved form and, therefore, highly mobile; trace metals like boron, zinc and cadmium have ratios of dissolved species to particulate species of between 2:1 and 1:1; copper, mercury, chromium and lead exhibit ratios of the aqueous phases to the solid phase between 1:2 and 1:4; iron, aluminium (and manganese under normal Eh conditions in rivers) are almost totally transported as solid particles. Sediment analysis, therefore, are particularly useful with respect to the less mobile elements, i.e., most of the heavy metals.

Although sediment analysis do not furnish quantitative data on the absolute degree of pollution, they can play a key role in ascertaining relative factors of enrichment whereby sources of pollution in the aquatic environment may be traced and monitored.

The Applied Geochemistry Research Group at Imperial College, London has applied geochemical reconnaissance by analyzing more than 50,000 sediment samples collected from England, Wales and Northern Ireland on an average of one sample per square mile to compile geochemical atlases for twenty elements (Thornton and Webb, 1973; Aston and Thornton, 1977). Such type of regional geochemical mapping is of immediate practical value in application to various environmental problems, such as those associated with agriculture, fisheries, pollution, public health and sludge dumping.

Fig. 2 illustrate the distribution pattern of arsenic and reflects the contamination from mining, urban and industrial sources (Aston and Thornton, 1975; Aston et al., 1975). On the basis of such mapping the authors have been able to highlight potential problem areas where anomalous high concentration of metal enrichment may result in the deterioration of potable water quality.

Aston and Thronton (1977) studied the composition of stream sediments and provided a useful and stable indicator of associated water. Bottom sediments collected from streams of both

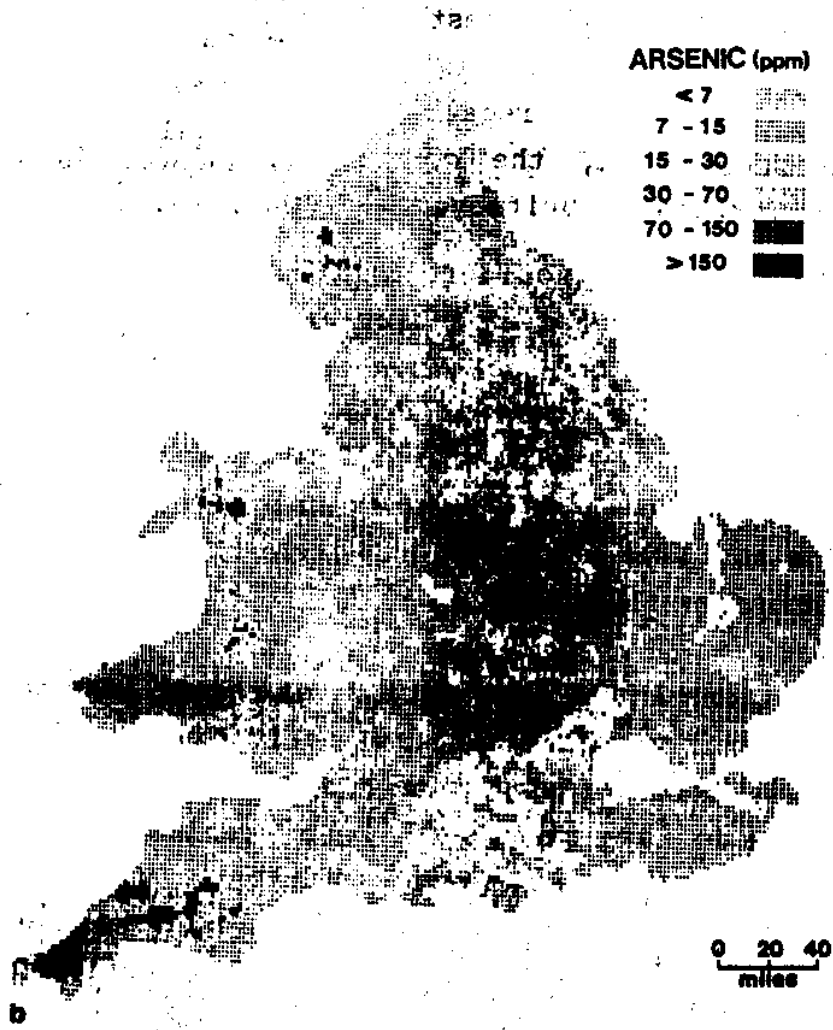


Fig. 2. Distribution of arsenic in stream sediments in England

mineralized (mining) and unmineralized areas were sieved through mesh to obtain $<200 \mu\text{m}$ fraction for analysis. The temporal variations observed in stream sediments were ascribed due to seasonal changes in bed loads resulting from (i) weathering fluctuations, and (ii) associated changes in grain size distribution.

The fine grained sedimentary deposits from suspended material in which the sorbed, co-precipitated, incorporated or otherwise bonded metal contents tend to accumulate, are particularly suited to such investigations. However, it is often overlooked that it is imperative to base such finding on a standardized procedure with regard to particle size, since there is a marked decrease in the content of metals as sediment particle size increases (Drifmeyer and Odum, 1975).

Sediment cores provide a historical record of events occurring in the watershed of a particular lake or bay and enable a reasonable estimate of the background level and changes in input over an extended period of time. This approach is especially valuable if the rate of sedimentation is known. Examples from lacustrine sediments cores have been given by Krishnaswamy et al. (1971); Ravera and Premazzi (1971); Ritchie et al. (1973); and Robins and Edgington (1975).

Variations in the stratigraphy of recent lake sediments were first performed to trace cultural activities on the rate of eutrophication by Murray (1956) from Wisconsin lakes in Northern Germany. Man made effects have been evaluated from the distribution of phosphorous and other nutrients (Livingstone and Boykin, 1962; Whiteside, 1965; Frink, 1967; Wentz and Lee, 1969; Duthie and Sreenivasa, 1971), from the distribution of iron monosulphide and in changes of diatom assemblages (Stockner and Benson, 1967). Pollen variations in recent lake sediments reflect historical changes in land use (Solomon and Kroener, 1971; Anderson, 1973; Kemp et al., 1974). Poon and Sheih (1976) found a close relationship between phosphorous and iron from sedimentary cores both in the lacustrine and marine coastal environments.

The foregoing discussion has clearly demonstrated that lateral variations in the chemical composition of surface sediments act as a guide to local pollution centers. Similarly, qualitative profiles of sediment data along a river course can be used to evaluate characteristic influences from industrial, municipal and agricultural sources, provided that the grain-size effects, which strongly influence the metal values, are taken into account.

Investigations on metal pollution by means of sediments commence with the determination of general hydrologic data such as, water temperature, pH-Eh values and conductivity and in the case of river systems, water flow rate at the time of sampling. For routine investigations on sediments, an analytic procedure is described in Fig. 3.

3.1 Sampling

Several devices have been developed for the extraction of surface sediments, sediment cores, interstitial water and suspended material. In most cases the particular apparatus serves only one purpose, as discussed below.

3.1.1 Soil and sediments

Sampling and storage of soils, rocks, minerals and dry sediments present problems than those encountered with water and biologic sampling material. For instance, the use of an ordinary spade for soil sampling is preferable to the use of an auger (Morrison and Pierce, 1974). Tools which are encased in teflon have been especially recommended from the inner part of the sample material which has not been in direct contact with the metal of the sampling device. Another important requirement is to observe the textural features of the sample and make a preliminary classification according to grain size. When sampling from beneath water coverage, great care must be exercised to leave the top layers undisturbed (Hakanson and Uhrberg, 1973).

3.1.2 Grab and core samples

Sampling from sediment below water coverage depends on the type of sample required. Coarse-grained consolidated material may be recovered, even from beneath overlying ice, by means of a mud-grab. This spring-loaded device is constructed from noncorrosive material and capable of extracting samples of 15 cm in diameter.

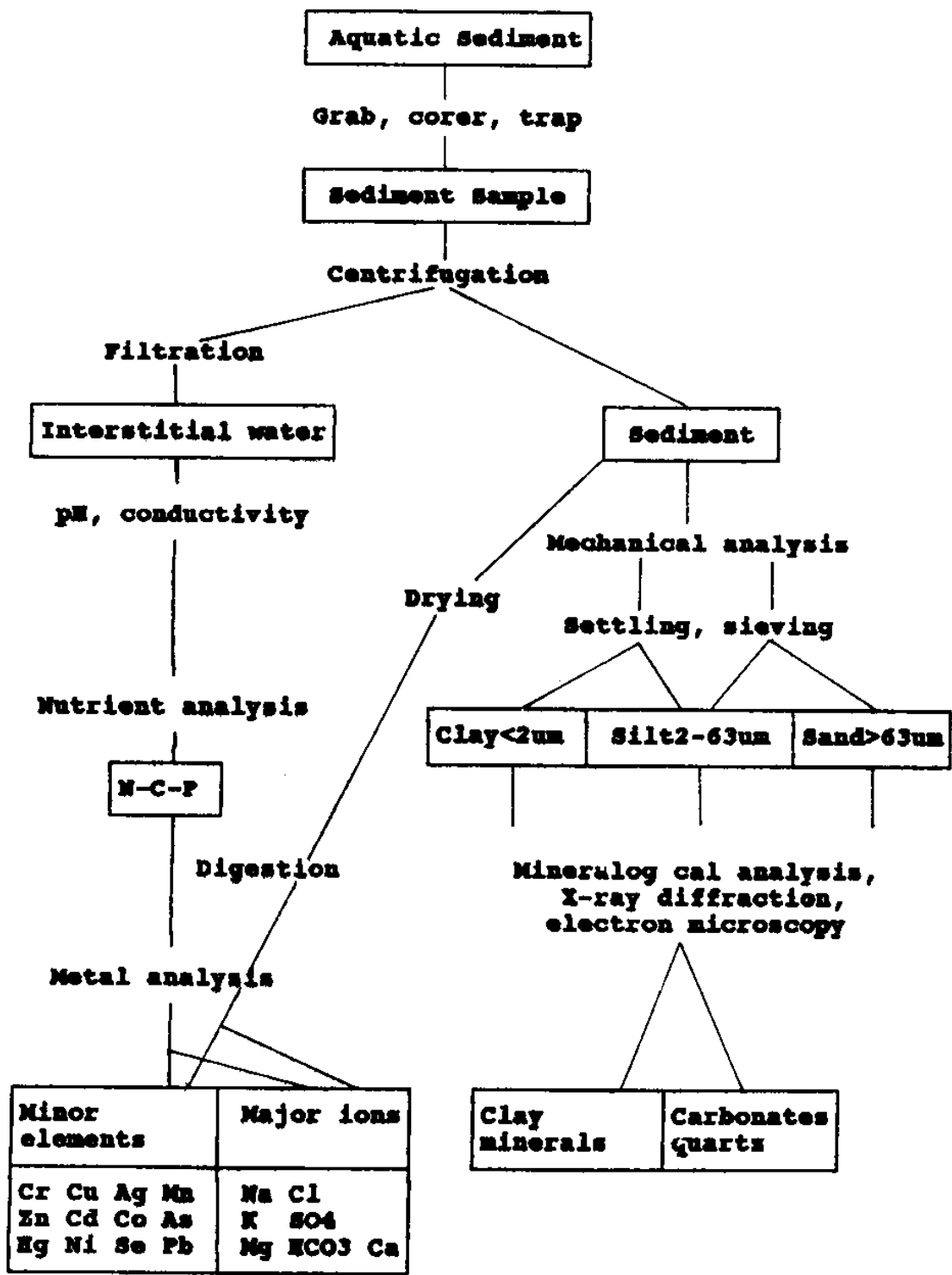


Fig. 3. Schematic sequence for analytical procedure in metal pollution assessment from sediments

Normally fine grained bottom sediments are collected with the aid of a sediment grab of the Van Veen type. The catch that keeps the two bowl-shaped sections of this dredger apart is released upon making contact with bottom sediment; withdrawal subsequently leads to a closure of the half sections and capture about 2 kg of sediment material with a penetration depth exceeding 20 cm.

If special importance is attached to collecting undisturbed sediment samples, the Ekman-Birge grab is most suitable. The grab is furnished with two flaps which are spring-loaded and enclose the sample material upon withdrawal. A modification of this grab after Lenz enables the subdivision of the sample into five profiles of 20 cm thickness by means of inserting horizontal plates.

The increased interest displayed in the reconstruction of historical developments of pollution influences has led to wide spread sampling of sediment cores. Such profiles may cover the last two hundred years of industrial development and in accordance with an average sedimentation rate for lacustrine and marine coastal environments of approximately 1 to 5 mm per year, which is applicable to moderately humid climates, the procedure entails sampling of a core of 20 cm to 1 m in length. Obviously, in areas where the annual sedimentation rate is much higher due to erosion, longer cores have to be extracted.

Gravity corers are used particularly in the study of sediment texture and evaluation of paleoclimatic changes for which cores of 2-3 m of fine-grained sediment material are required (under favorable conditions cores of 10 m can be recovered). A major disadvantage arises from the fact that the top section of the core, which is of particular interest in pollution studies, is often lost.

3.1.3 Bottom sediment traps

These have been used during the last more than 50 years for the determination of sedimentation rates. Now a days many

techniques are available which include bottles, funnels and pots suspended from cables. Bottom sediment traps have been constructed even for sandy river deposits. Recent developments have been described by Hakanson (1976), a new bottom trap having been introduced. The bottom plates consists of Plexiglas which is roughened to increase friction; brass arms with lead weights increase the stability and eliminate disturbances in the system.

3.1.4 Suspended materials

There are recovery methods which can register suspended material contents as low as approximately 1 mg/L. Muller et al. (1976) described a high-pressure filtration system in which 8 liters of river water are filtered through a 0.45 μm membrane filter within approx. 20 min. These filters are constructed from stainless steel and are connected in pairs; the second filters is silver-coated to permit direct analysis by X-ray diffraction techniques. Other methods for the recovery of particulate matter are continuous-flow centrifugation and suspended sediment traps, which are immersed into the flow medium.

3.2 Storage

The storage of the solid material is subjected to stringent conditions. Usually air-drying should not have much effect on the total trace element content, but if speciation, organic extractable trace elements, etc. are of interest, any procedure of drying may lessen the validity of the sample analysis (Maienthal and Becker, 1976). For instance, air-drying of the sample markedly affects cation exchange capacity and iron speciation, and appears to decrease the pH slightly (Schalscha et al., 1965). Drying and freezing may result in irreversible change which will also affect the complexation state of the trace metals (Hesse, 1971). In the opinion of many authors freezing at -20° to -30° C is the best method of storage.

3.3 Mechanical Analysis

Mechanical analysis normally begins with the sieve

procedure, the separation of the sand fractions ($>63 \mu\text{m}$) from the finer-grained sediment particles. These are then separated into silt and clay fractions by allowing the heavier particles to settle using water as suspension medium. This method, however, can cause a deceptive contamination of the sediment due to metal corrosion of the sieve wall and base. For this reason, it is preferable to place all samples irrespective of grain-size in Atterberg settling tubes and separate the $<2 \mu\text{m}$ fraction by repeated suspension and settlement cycles (five to ten times). This is followed by a separation of the increasingly coarser silt fraction.

3.4 Mineralogical Analysis

The main aim of the mineralogical analysis is to determine the origin of the individual sediment component; this is generally done by an investigation of the distinct grain-size fractions. The clay minerals are determined from the fraction $<2 \mu\text{m}$ previously freed of most interfering organic substances by hydrogen peroxide treatment. The sample is disintegrated with ultrasonic waves and suspended onto a glass slide and subsequently run on X-ray diffractometer.

The carbonate determination are carried out in each fraction. The total carbonate content is generally determined by the treatment with hydrochloric acid.

3.5 Sediment Digestion

The digestion of the sediment sample is carried out by one of the following methods:

3.5.1 Hydrofluoric acid decomposition

The procedure which is usually followed for dissolving sediments for spectroscopic analysis employs either sulfuric acid or a mixture of nitric acid and perchloric acid with hydrofluoric acid. Decomposition of the material is effected by reaction of hydrofluoric acid with silica forming gaseous silicon

tetrafluorides. As the evaporation proceeds, most of the metal fluorides are converted to sulfates or perchlorates which in turn can be dissolved by hydrochloric acid. The HF-H₂SO₄-HNO₃ decomposition permits the ready extraction of the hydrofluoric acid residue, and the acid mixture can then be handled more safely; a definite disadvantage is the possible formation of sulfates which can be dissolved only with great difficulty. The advantage of the HF-HClO₄ decomposition is that the perchlorates, with the exception of K, Rb and Cs compounds, are more soluble. If the substances to be analyzed contain bituminous or other organic compounds, they must first be oxidized by heating in order to reduce the danger of explosion; this can also be achieved by adding nitric acid to the substance. The following procedure for sediment samples is used: Weigh 250 mg of the sample material in a platinum crucible and evaporate to dryness with a mixture of 5 mL HF and 2 mL cm HNO₃ on a sand bath. Repeat evaporation with 2 mL HNO₃. Dissolve the material with 5 mL dilute HNO₃ (15%) by heating for 30 min and transfer to a 50 mL volumetric flask.

3.5.2 Hydrochloric-nitric acid (Aqua regia) decomposition or digestion by nitric acid

This method is normally used for the determination of more volatile elements, e.g., mercury, arsenic and cadmium. Transfer 50 mg of sample material to a 50 mL measuring flask and treat with 3 mL HNO₃-HCL (1:3) on a sand bath for 30 min at moderate temperature (60° C). Allow for cooling to room temperature and dilute with distilled water to a 50 mL. Alternatively, PTFE-bombs are used for digestion of sediment samples to be analysed for Cd, Hg and As: 100 mg of powdered sediments are weighed into the teflon-bomb and 5 mL of aqua regia is added. The sealed bomb is heated at 110° C for 2 h in an oven.

3.5.3 Lithium metaborate fusion (with simultaneous determination of silica)

Place 50 mg of sample material in a platinum crucible and mix with approx. 200 mg LiBO₂ before heating for 15 min at a

temperature of 1100° C; chill the molten matter. Add 25 mL of 10% hydrochloric acid and dissolve the entire mass using a magnetic stirrer. Transfer quantitatively and dilute to 50 mL in a volumetric flask.

3.5.4 Transfer of solid suspension into graphite cuvettes

Trace metals in solid material can be determined without digestion provided the particles are sufficiently fine grained to ensure homogeneous distribution in suspension. This prerequisite is always met by the $< 2\mu\text{m}$ fraction. Numerous investigations using sediment analysis in pollution control have shown that it is unnecessary to obtain full digestion of all sediment components, including metals bound into the internal structures of silicates and other detrital minerals, since the pollution effects usually occur at the surface of the sediment particles and in the autochthonous precipitates. Anderson (1974) has therefore suggested to a simple HCl-HNO₃ (1:1) digestion for the determination of heavy metals by atomic absorption spectrometry.

4.1 Geochemical Reconnaissance of Aquatic Sediments

Geochemical investigations of stream sediments have long been recognized as a standard practice in mineral exploration. The method of stream sediment sampling is preferentially employed in remote areas in order to obtain a preliminary idea of the possible mineralization zones (Boyle et al., 1955; Hawkes et al., 1957). By more extensive sampling and analysis of the metal contents in water, soils and plants, the probable enrichment zones can be narrowed down and, in some cases, localized as exploitable deposits (Hawkes and Webb, 1962).

In stream sediment sampling for geo-prospecting, Nicol et al. (1969) have shown that with statistical and mathematical models it is possible to reduce the number of variables; examples of factor analysis of stream sediment geochemical data are given by Saager and Sinclair (1974) from the Mount Nansen area, Yukon Territory, Canada. For different mining environments of mid-Wales and Colorado, Wolfenden and Lewin (1978) have found systematic relationship between metal concentration and particle size and distance downstream. Examples of practical application have been given by Symader and Thomas (1978) for the Red Main River in Germany. Synoptic graphical presentation of correlation coefficient matrices were used by Davaud (1977) to demonstrate the differences in chemical association in recent and older sediments from Swiss lakes. Wolfenden and Lewin (1977) explained how to produce a three dimensional view of lead values in sediment samples from a meander loop of River Rheidol in mid-Wales, which is polluted by mining wastes.

The dispersion of metals into lake sediments was depicted by Allen et al. (1974) in the study 'Mercury and arsenic levels in lake sediments from the Canadian shield'. According to the authors, the fate of metal ions, originally derived from chemical weathering and mechanical disintegration of host rock is controlled by many factors involving atmospheric precipitation, water movement, soil movement, change in redox and

pH conditions, absorption-desorption processes, chemical complexation, precipitation and hydrolysis, uptake by and decay of vegetation and biochemical-bacterial interactions.

A large proportion of the fluvial transport of matter occurs in the form of suspended material with mean grain size of 2-63 μm (silt grain size of 10-20 μm are very common). During a decreased rate of flow, i.e., if the total load exceeds the available energy, the suspended material gradually sinks to the bottom and is deposited in the river bed where it is partially incorporated into the bottom sediment. The particles with larger mass (and correspondingly coarser grains) are the first to be deposited, followed by the increasingly lighter, finer-grained components. The fact that these deposits partly remain intact in spite of any subsequent increased rate of flow is due to specific cohesion characteristics of the fine-grained particles.

The distribution of metals in the grain-size spectrum has been studied widely. Gibbs (1977) formulated calculations of the mass transportation of various elements and characteristic metal association with particulates in relation to the grain size spectrum. It is obvious that grain size exercise a determining influence on the metal concentrations, not only with regard to the samples selected for pollution control but also to the natural background data. This fact must be taken into consideration particularly with respect to studies of river sediments where variations in grain size are usually large.

Several procedures have been adopted to minimize the grain size effects on trace metal data. De Groot (1971) has used an extrapolation method for correcting the metal analysis data for the behavior of mercury and other metals in the suspended matter of the highly polluted river Rhine in the Netherlands. A similar approach was used by Renzoni et al. (1973) for the assessment of mercury pollution from a large chloralkali factory on the Tuscan coast of the tyrrhenian sea. Repeated analysis of mercury in the bottom sediments showed that the mercury is strictly related to the average diameter of the sediment grains.

In many studies on metal pollution in river sediments, the clay/silt fraction (and in some cases the fine sand fraction as well) is separated with nylon sieves in order to reduce at least the strongest grain size influences. Examples for the use of the $< 200 \mu\text{m}$ fraction can be found in Copeland's (1972) study in the lake Michigan Environment and in Thornton et al.'s (1975) ambitious study of the geochemistry of sediments from rivers and estuaries in Great Britain. Others working on sediment analysis prefer to use the $< 63 \mu\text{m}$ fraction (silt/sand boundary). This latter method was used in the exploration studies undertaken with the help of river and lake sediments by the Geological Survey of Canada (Cameron, 1974). Chester and Stoner (1975) used the $< 61 \mu\text{m}$ fraction which were separated by wet sieving with re-distilled water through nylon sieves.

4.2 Natural Metal Content

Trace metals in recent sedimentary deposits can generally be divided into two categories: lithogenic and anthropogenic (Hellmann, 1970), often generally referred to as geochemical and man-made respectively. Metals such as zirconium, rubidium and strontium, which are derived from rock material by natural weathering processes, constitute the first group. The second group is made up of metals which have become enriched chiefly as a result of man's activities, and includes among others chromium, cobalt, nickel, copper, zinc, cadmium, mercury and lead. Between these two groups there are combinations; for example, the enrichment of mobile elements such as manganese and iron, which may well have had civilizational origins such as extreme eutrophication. By their own accumulation, these metals can cause other elements to accumulate.

The practical experience of the last few years has shown that positive proof of pollution sources can best be obtained by means of sediment analysis near specific sources, particularly at points where industrial sewage is discharged into surface water systems. Additional difficulties arise in tracing specific influences of metal pollution from mixed sewage systems, particularly where municipal water predominates. There are other

metal enrichments, which simply cannot be pinpointed to a specific supply source and which are consequently described as resulting from diffuse or nonpoint sources. They may be locally included in the background value in order to obtain easier evaluation of the definite or point sources in a particular area.

Typical nonpoint sources for elevated metal concentrations are represented by the runoff from agricultural land and by atmospheric precipitations. Sheet erosion or seepage may, for example, transport higher concentrations of cadmium and arsenic, resulting from fertilizers or sewage sludge, into adjacent waters, both in solution and as solid particles. Lead from atmospheric precipitations can also reach surface waters in a similar way: on the one hand by mechanical erosion of soil particles which are transported in the form of fluvial particulates and deposited in inland water systems and marine basins, and on the other hand from the chemical processes of leaching and mobilization of the heavy metals from particulates in either a dissolved or complexed form and a reprecipitation onto organic and inorganic sediment particles in a depositional basin. In densely populated areas a considerable amount of atmospheric precipitation with its metal load enters the surface waters via the public sewage system, thus rendering a clear differentiation between the influences of direct sewage and atmospheric heavy metal contamination impossible. These various interactions must be considered when pollution assessment is undertaken by sediment analysis. In the following sections, a number of examples will outline the possibilities, as well as the limits, of these methods.

4.3 Metal Pollution in Lake Sediments

The significance of sediments in the assessment of the quality of aquatic systems is well illustrated by examples from the lacustrine environment. The reason for this lies in the fact that natural fresh water lakes have been the centers of important cultural developments since earlier days of civilization. As a consequence of increased population and industrialization densities, the threat of pollution has become most acute in areas

dependent on natural and man-made lakes as a source of potable water.

The first observation of changes in the lacustrine environment were made on lake Zurich. Hutchinson and his team (1943) and Zullig (1956) drew attention to a major application of geochemical research in the study of recent lake sediments. Since sediments may be regarded as a response of the conditions in an aquatic system, the study of freshwater deposits could play a key role in ascertaining the complex hydrochemical interactions which take place in lakes and rivers. During the last three decades sediment analysis has acquired a new dimension by being employed as a tool to trace man-made pollution influences in inland and coastal waters.

Mining activities are often associated with higher metal levels in the environment. Allen (1974) studied the distribution of nickel in sediment cores of nickel mining area in and around Sudbury basin. Apart from nickel contamination around Sudbury, the effect of arsenic at Red Lake and of copper at Chibougamau were also investigated. The cores were taken from the center of the lakes (as the center is most likely to reflect the overall variation within the lake in relation to man's activities). Most of the lakes had accumulated 10 cm of sediments within the last hundred years. Thus samples of cores taken from below this depth reflected the natural levels which occurred prior to ore exploitation, whereas samples within the upper 10 cm level has expected to indicate the extent of subsequent mining activities.

Lake sediment cores have been used to obtain a historical picture of the progressive increase in metal content as a result of natural and artificial phenomena (Kobayashi et al., 1976). Several authors showed that sediments from reservoirs located downstream from heavy metal mining and milling areas contain higher concentrations than reservoirs in other areas (Pita and Hyne, 1975; Gale et al., 1976). Hardin and Whitton (1978) suggested that high levels of heavy metals in the sediments of lakes and reservoirs depending on metal rich inflow

waters could be widespread in Europe and indicated Sardinia among the exposed areas. Schintu et al. (189) studied the total concentration of metal ions in sediments and their re-mobilization in the water.

Wittmann and Forstner (1975) described the pollution effects with regard to metal pollution in lake sediments of South Africa. The high zinc and lead values were ascribed due to domestic waste effluents, while cadmium and mercury due to industrial discharges (Wittmann and Forstner, 1976).

Wentsel and Berry (1974) reported very high cadmium content in western part of the Palestine lake, Indiana due to discharge of electroplating waste via a small ditch, whereas the eastern part affected only to a small extent. Nightingale (1975) studied the accumulation of lead in the first few centimeters of soil in California.

Several studies on metal investigations in sediments were carried out in Sweden (Bengston and Fleischer, 1971; Hakanson, 1973, 1974, 1976; Jernelov and Lann, 1973, Hakanson, 1977). Other studies on lakes in Finland have been done by Hinneri (1974) and Sarkka et al. (1978). Switzerland has also been the scene for a large number of studies concerning the burden of heavy metals in sediments of lakes (Vernet and Thomas, 1977a,b; Blackburn, 1973; Davaud, 1977). Metal pollution was reported by Stern and Forstner (1976) and Molnar et al. (1978) from lakes and dams in Slovenia, Yugoslavia. The investigations of trace metal contents in lake sediments from the Federal Republic of Germany were carried out by Forstner et al., 1974; Forstner and Muller, 1974 and Muller et al., 1977.

Investigations on lake Biwa in Japan (Nakamura et al., 1974; Satake et al., 1975; Tatekawa et al., 1975) have shown local heavy metal enrichments that are more probably attributed to rapid industrialization, urbanization and land development. Stronger contamination by zinc, lead and cadmium was found by Goldberg et al. (1976) in sediment cores from the Palace Moat, Tokyo.

Investigations of the vertical distribution of trace elements in sedimentary cores clearly indicate the increase of metal pollution during the last 100 to 200 years. A general increase in the concentration of various elements was observed in the lakes studied by various researchers. Various examples of metal contamination recorded in dated sedimentary cores have been described in detail by Forstner and Wittmann (1983).

One of the most serious forms of environmental pollution is the contamination of lakes by mercury wastes. This is due to the specific toxic behavior of mercury, arising from the biosynthesis of mercury-alkyls and to a distinct residue-forming tendency of these substances in aquatic ecosystems. Mercury concentrations in sediments represent a particularly good indicator for various forms of cultural activities, e.g., from agricultural use of fungicides, atmospheric fallout from power plants, influences from mining and smelting processes, sewage input etc. Sediment cores from lakes exhibit a characteristic increase of the mercury concentrations. One likely source of mercury accumulation in remote areas is particulate matter, either from industrial plants burning fossil fuel, or from the smelting of copper, lead and zinc ores (Mathis and Kevern, 1973).

Sediments from lake Saoseo and lake Tuma in southern Switzerland are contaminated with mercury of atmospheric origin (Vernet and Thomas, 1972). The study of Clay lake in Canada was conducted by Armstrong et al. (1972) and Armstrong and Hamilton (1973). mercury distribution in the sedimentary profiles from the central parts of the lake show concentration of more than 8 ppm in the upper 4 to 6 cm of the cores, whereas mercury content below 6 cm was usually less than 1 ppm.

4.4 Metal Pollution in River Sediments

The significance of suspended and bed sediments in aquatic environment has long been recognized. They are also important vectors for the transport of contaminants in river systems (Allen, 1986; Ongley et al., 1988). A number of studies on river sediments have been conducted under both scientific

aspects and direct pollution control. These investigations cover a wide range of regional peculiarities and indicate municipal, industrial and agricultural influences over a wide spectrum of pollution intensity, which can comprise three to four orders of magnitude for a specific metal contaminant.

Mineral exploration based on the distribution of ore metals in stream sediments has become a generally accepted method of mineral reconnaissance. Mercury and its compounds play a important role in geochemical reconnaissance in stream sediments. Characteristics enrichment of toxic metals from mining operations and natural sources also occur in fluvial sediments. Batti et al. (1975) found 500 times mercury concentrations than the background values in river deposits from the Mt. Amiata region, Tuscany, Italy. A review on mercury contamination in rocks, soils and stream sediments of the United States was prepared by Pierce et al. (1972).

Laszlo et al. (1977) conducted studies on Sajó river in Hungary. Analysis of pelitic fractions ($< 2 \mu\text{m}$) have been used by the Institute for Sediment Research at Heidelberg University to assess the most significant centers of heavy metal pollution of the major rivers - Danube, Rhine, Neckar, Main, Ems, Weser and Elbe. Investigations on suspended material and sediments of the Hudson River between Kingston and Manhattan, carried out by McCrone and Koch (1966), revealed signs of metal pollution by industrial and municipal effluents.

Turekian and Scott (1967) studied the concentrations of chromium, silver, molybdenum, nickel, cobalt and manganese in suspended material from eighteen rivers in the southern and eastern United States, and were able to determine a distinct accumulation of Ag, Ni and Co, particularly in the case of the Susquehanna River, which they ascribed to industrial contamination. A comparison of the values obtained with the naturally occurring concentrations led the authors to conclude that "the annual transport of trace elements by the Susquehanna River as suspended material is sufficiently large to be of possible economic interest if the demand for some of the metals

increases and the metals are present in an easily reversible form".

Forstner (1980) compiled cadmium concentrations and sources in sediments for some European rivers (Table 2) and reported that due to different sample preparation techniques the data are not directly comparable. Various studies on mercury pollution in river sediments performed during 1970's in different areas of North America have been compiled in Table 3.

Geochemical investigations on selected sediment cores from the Kingston Basin, Upper St. Lav River (Johnston, 1977) revealed surface enrichment in Cd, Co, Cu, Ni, Pb, Zn, Hg, and As; mean values, which can to some extent be considered as background data for polluted areas, were evaluated at Cd = 0.70, Co = 13, Cu = 26.7, Ni = 41.5, Pb = 28.5, Zn = 120, and As = 3.5 ppm. Various studies were performed on the metal contents in the Ottawa River sediments (Oliver, 1973; Oliver and Agemian, 1974). Of particular interest here are the distribution and bed movement of mercury pollutants (Kudo and Hart, 1974; Rust and Waslenchuk, 1974; Townsend et al., 1974; Kudo et al., 1975, Waslenchuk, 1976). The transport mode of copper and nickel was investigated in the Wanapitei River, which flows through highly metal-contaminated areas near Sudbury (Fitchko and Hutchinson, 1977).

The results of geochemical surveys on mercury levels in the rivers of western Canada, during the period of 1970-1976, have been published by Canada Inland Waters Directorate, Ottawa, (1976), and include sediment analysis data. Trace metal investigations on the sediments of the lower Fraser River and estuary indicate moderate contamination, mainly resulting from municipal sewage effluents (Hall and Fletcher, 1974; Grieve and Fletcher, 1976).

A comparative study of heavy metal concentrations in river mouth sediments around the Great Lakes (Fitchko and Hutchinson, 1975) indicated the highest mercury discharge into the lake Ontario arising from the Niagara River, followed by the

Table 2. Cadmium concentrations in sediments of rivers and streams in Europe

	Cd Conc. (ppm)	Reference
Elbe River, F.R.G.	2.9-19.4 max.20	LichtfuB and Bruimmer (1977)
Sajo River, Hungary	max. 20.0	Literathy and Laszlo (1977)
Blies River, Saar/F.R.G.	< 0.5-24.0	Becker (1976)
Voglajna River, Yugosl.	max. 66	Stern and Forstner (1976)
River Conway, UK	3-95	Thronton et al. (1975)
Stola River, Poland	max. 116	Pasternak (1974)
Neckar River, F.R.G.	max. 230	Forstner and Muller (1973)
River Tawe, Wales	max. 355	Vivian and Massie (1977)

Table 3. Mercury concentrations in sediments of rivers and streams in North America

	Conc. (ppm)	Reference
St. Lawrence River	0.18	Johnston (1977)
Oswego River	0.67	Fitchko and Hutchinson (1975)
Ottawa River	3.0	Rust and Waslenchuk (1974)
Islais Creek (Calif.)	6.9	Smith (1972)
Stream in Taylor Mts (Alaska)	10.0	Clark et al. (1970)
Tennessee River	32	Derryberry (1972)
(North Fork)		Turner and Lindberg (1978)
(Wheeler Dam)	(550)	Lloyd et al. (1972)
Wabigoon (Ontario)	56.0	Buffa (1976)
St. Clair River	60.0	Cline et al. (1973)
	(1700)	
Detroit River	86.0	Turney (1971)
Wisconsin River	792.0	Iskandar et al. (1972)

Oswego River; core samples revealed 5 to 15 fold higher concentration of lead, cadmium and chromium in the sediments of the Milwaukee River in comparison to material from the mouth of the Raisin River, both draining into Lake Michigan. Several geochemical surveys on river sediments have been conducted in Wisconsin, Illinois, Indiana and Michigan (Hess and Evans, 1972; Konrad, 1972; Mathis and Keern, 1973; Romano, 1976); trace metal concentrations of copper, zinc and cadmium in the upper Peoria lake sediments (Collinson and Shimp, 1972) were 8, 12 and 26 times higher than the respective levels in the sediments of the Illinois River. Similar comparison of the concentrations of lead, cadmium, zinc, copper and cobalt in streams and lakes of the Cayuga Lake Basin, New York, indicated that the levels of these elements, in suspended particulates carried by the streams flowing through Ithaca, New York, were higher than in streams flowing through rural area due to direct result of the impact of urbanization (Kubota et al., 1974). Although there is no characteristic source of metal pollution in the watershed of the Seneca River, New York, industrial and sewage effluents produced local concentrations in excess of safe levels for waters that are fished (Blackburn et al., 1977); the Genesee River, which also drains into Lake Ontario, appeared relatively unaffected by metal contaminants (Reddy, 1977).

A study of cadmium and nickel concentrations along the east side of the Hudson River in New York State indicated further evidence of considerable pollution, which could have resulted from battery plants, particularly from those employing cadmium (Kneip et al., 1974); a sample close to the discharge plant contained approximately 40,000 ppm cd, another sample 100 m downstream indicated 2100 ppm cd. The amount of cadmium in the sediments of the Foundry Cove disposal site from these discharges was estimated to be approx. 20-50 tons; core sediment showed a maximum of more than 250 ppm cd at the top and approx. 2 ppm cd as background (Bower et al., 1978).

Several metal pollution surveys have also been conducted on rivers which drain the east coast of the United States. Two studies dealing in particular with the geochemistry

of river sediments were conducted on the Murderkill and St. James River in the Delaware Coastal Plain (Bopp et al., 1973) and in the lower reaches of the Potomac River (Fauth and Houser, 1973). In the Mississippi drainage system the Tennessee River gained particular attention due to the various characteristic pollution sources from industrial and municipal centers and, most important, from mining activities.

Several comprehensive studies were performed on the metal distribution in bottom sediments, suspended particulates and water of the northern Tennessee zinc district and its effect on downstream metal concentrations (Derryberry, 1972; Perhac, 1972, 1974b; Perhac and Whelan, 1972; Hildebrand et al., 1975). From the western tributaries of the Mississippi, sediment reconnaissance studies have been performed on several streams in Kansas (Angino et al., 1974), on Skeleton Creek/Cimarron in Oklahoma (Namminga and Wilhm, 1977) and on the Buffalo River in Arkansas (Bowen and Steele, 1975; Steele and Wagner, 1975). The highest lead concentrations (> 200 ppm in Deep Fork River) occurred in sediments from the more densely populated areas with heavy vehicular traffic. Mining activities in the New Lead Belt of Missouri, the world's largest lead-mining district, still pose some problems for water quality despite modern processing methods (Jennett, Wixson and colleagues, 1973-1977), as there is obviously a considerable metal transport by fine particulates (Jennett et al., 1973).

Geochemical studies in the lower Mississippi revealed no characteristic increase in the heavy metal content of the sediment load (Hartung, 1974); the minimum annual transport rates of metals in particulates were estimated at approx. 2000 tons for arsenic, 85 tons cadmium, 1400 tons chromium, 3500 tons lead and 84 tons mercury, based on an annual sediment load of 256×10^6 metric tons. The impact of man's activities is well illustrated by the fact that the cadmium discharge rates in the Mississippi River are considerably lower than the cadmium output from the combined sewage effluents of Los Angeles and New York in the delta sediments of Mississippi River (Trefry and Presley, 1976; Trefry, 1977).

In Europe, the assessment of river pollution by means of sediment analysis was initiated in the 1960's in the Netherlands by De Groot and associates. Their studies centered on investigations on the transport of sediments from the Rhine and Ems Rivers into the North Sea and on the behavior of trace metals in the mixing zone of freshwater and seawater (De Groot, 1966; De Groot et al., 1971, 1973; Van Driel and De Groot, 1974; Salomons and De Groot, 1977). At the same time in the Federal Republic of Germany, Hellmann and co-workers (1970-1973) studied the Rhine River and its tributaries in detail to ascertain the extent to which the process of water reclamation from bank filtrates was being endangered by increasing river water pollution.

In order to establish the origin of troublesome mud deposits, special emphasis was given to the role of zinc as a characteristic element in domestic and industrial waste effluents. A particularly heavy enrichment of zinc (up to 1% in dry sediment) was noted in the Ginsheimer Altrhein, a cut-off channel of the Rhine River in the vicinity of Mainz (Hellmann, 1970). Laskowski et al. (1975) revealed unusually heavy enrichments of cadmium, silver and mercury.

A general survey on the metal concentrations of the pelitic sediments of major rivers in the Federal Republic of Germany indicated that the lower section of the Neckar River is highly polluted with cadmium suspecting direct poisoning of the river water. Further analyses of water samples taken at a distances of 1-2 km confirmed these fears. Although this very high concentration was diluted by the water of the Neckar (having Cd-contents of less than 1 $\mu\text{g}/\text{l}$), elevated Cd-concentrations was found all the way downstream to the point where the Neckar River flows into the Rhine. The unusual increase in cadmium, which among other things led to fish poisoning, was caused by a pigment dye production plant which released an annual total of approximately 10-20 tons of cadmium in dissolved or particulate form into the river.

Houba et al. (1983) studied the various factors

affecting the concentration of metal ions in the sediments of the Vesdre river in Belgium. The concentration of trace elements discovered in recent sediments of the river and its tributaries have been reported by Houba and Remacle (1982). The concentration of metal ions were found 5 to 10 times higher than the reference levels.

Metal pollution in rivers in Switzerland were investigated by Vernet and co-workers (Vernet and Johnstone, 1974; Vernet et al., 1977). In the Rhone River, upstream from Lake Geneva, an important contamination of mercury has been located around Monthey, originating from anthraquinone and chlor-alkali plants. With regard to other trace metals it has been found that the dominance of Cr in Rhone sediments, which is associated with Sn in the upper reaches and with Zn downstream, can be ascribed to a number of chromium-plating industries. The Orbe basin in western Switzerland is characterized by the association of Zn and Co. The main association in the polluted sediments of the tributaries of Lake Morat is Cr-Pb, with one occurrence of Cd reaching 30 times the background level. The Aar River upstream from Solothurn (northwest Switzerland) is characterized by Zn-Cu pollution, with a major source at Thun. Finally, chromium, zinc, and tin are strongly enriched at the junction of the Aar with the Rhine. Serious cases of heavy pollution have also been recorded in other European countries, the most critical of which are due to elevated mercury and cadmium concentrations.

In Sweden, the mercury load in fish, water and bottom sediments has been reported by Hasselroth (1968). Similar studies in North American lakes and rivers are reported by Tumey (1971). Maximum accumulations of mercury in river sediments were found in Ume Alv, downstream from the Bowaters wood-pulp mill, where in 1967 the mercury levels measured 100 ppm. Berndtsson (1990) studied the transport and sedimentation of pollutants in a 5 km long reach of river Hoje in south of Sweden and reported that half of the transported zinc is retained in the stream sediments.

Sediment reconnaissance studies on heavy metals have

been reported in Poland (Pasternak and Glinski, 1969, 1972; Pasternak and Antoniewicz, 1970, 1971; Pasternak, 1973). Stream sediment data indicate particularly heavy enrichment in the vicinity of mining and smelting centers, e.g., at the Miasteczko Slaskie zinc plant in south Poland, where up to 120 ppm cadmium and 5000 ppm zinc were determined in the Sztota stream, a tributary of the Mala Panew River (Pasternak, 1974).

Very little work has been published on the metal pollution in rivers of the U.S.S.R. One of the few reports is from Liperovskaya and Drozhbina (1972) and concerns the pollution of Moscow River muds and their effect on the distribution of oligochetes. The heaviest pollution was recorded just downstream from Moscow city proper, possibly as a result of purification plant outflow near Nagatino and Perewa. It was further reported that after the river bed was cleaned in winter 1969/70, data from 1970 show a distinct decrease in copper contents in the sediment.

In Austria, heavy metal studies center mainly on the Danube (Rehwooldt et al., 1975). The mercury contents of the Danube sediments were found to be locally enriched; this appears to be partly due to the sediments consistence and current phenomena. Copper mine tailings contribute greatly to the pollution in the lower reaches of the Salzach River, a tributary of the Danube.

Micropollutants have been studied in the Hungarian section of the Danube since 1970 (Szebelledy and Literathy, 1970); sediment analyses indicated that there is no characteristic increase of mercury in the Danube from the Austrian/Hungarian border to Budapest (Literathy, 1974). Heavy enrichments of cadmium and mercury were observed in the sediments from the Sajo River in northeast Hungary (Literathy and Laszlo, 1977).

River sediments from the area around the Sava in Slovenia (Yugoslavia) indicate the background values of samples from the upper course of the river, as well as local current enrichments to be due to specific influencing factors (Stern and

Forstner, 1976). A sample from the industrial area of Celje (a tributary of the Savinja) was found to contain 1.5% zinc and revealed enrichments of Cd, Cr, Pb and Cu, which were 40 to 200 times higher than normal.

In Serbia (Yugoslavia) drainage water and stream sediments below the cinnabar mine at Mount Avala near Belgrade have an abnormally high content of mercury and arsenic (Maksimovic and Dangic, 1973). The cinnabar ore contains abundant iron sulfides and the acid solutions thus produced (pH = 2-2.6) dissolve large quantities of trace elements available in the altered serpentine minerals and in ore. Although the arsenic contents in the sediments show a steady decrease from the mine to the merging of the Potocni into the Topciderska River (a distance of 5 km), mercury contents exhibited irregular distribution. The authors explained this development as a result of the transport of mercury on Fe-hydroxides in the form of very fine-grained cinnabar particles with the mine effluents.

Rivers in Britain have been protected from high levels of industrial contamination by strict legislative control on discharges. Trace metal pollution is principally restricted to estuaries and a narrow coastal margin (Jaffe and Walters, 1975). On the mainland itself, however, areas do exist where natural weathering processes, together with past and present mining activities, have given rise to high levels of some metals (Cu, Pb, Zn, Cd, Sn and As) in sediments and waters draining these mineralized areas.

The rivers in southwest England are highly polluted by metals such as arsenic (>5000 ppm in the River Tamar), lead (4400 ppm in the River Gannel), copper (6500 ppm in the River Red) and zinc (7000 ppm in the River Camon). Intensive regional geochemical reconnaissance surveys in the assessment of water quality have been undertaken by the Applied Geochemistry Research Group at Imperial College, London (Webb et al., 1968, Thornton and Webb, 1970, 1973, 1975, 1977; Elderfield et al., 1971; Aston et al., 1974, 1975; Aston and Thornton, 1975, 1977; Colbourne et al., 1975; Thornton et al., 1975; Thornton, 1977).

Stream sediment geochemistry also revealed heavy pollution in Israel's inland water. Studies on the Qishon River, which, following the Jordan, is Israel's largest river system, were commenced in 1973 (Kronfeld and Navrot, 1974, 1975) and were centered mainly on the tributary of the Nahar Gadura, which is solidly lined by numerous industrial enterprises, including metal works, chemical factories, battery and electrode plants. Cadmium levels are reported exceptionally high, attaining more than 120 ppm.

Japan, together with North America and Europe, is one of the areas in the world most severely affected by environmental pollution. Here, it is mainly the use of catalysts and the production of plastic stabilizers and pigments which, in many areas and especially in coastal regions, has led to the dangerous increase of metal levels in sediments. Stream sediment studies revealed high concentrations of mercury (up to 18 ppm) and cadmium (40 ppm) in the Tama River near Tokyo (Goto, 1973; Suzuh et al., 1975, 1976).

Sakai et al. (1986) studied the distribution of heavy metals in water and sieved sediments of the Toyohira river in Japan, polluted by municipal, industrial and mining effluents. Terashima et al. (1979) have studied the mechanism of transport of heavy metals in water to bottom sediments. Further studies on the mechanism of distribution and transport of trace adsorptive pollutants in river have been reported (Terashima and Nagayama, 1979; Terashima and Tsukamoto, 1980).

In the sediments of the irrigation ditch of a Braun tube factory situated in the Ibaraki Prefecture northeast of Tokyo, concentrations of cadmium up to 368 ppm were detected (Asami, 1974). Heavy metal pollution in river sediments were analyzed from 85 points in the Chiba Prefecture southeast from Tokyo (Nakajima et al., 1976). The influence of an arsenic mine in the Gifu Prefecture (North of Nagoya) on the Wada River was studied by Kato et al. (1973); the mine sediments contained approximately 6% As and the river sediments 500-2400 ppm As. Azumi and Yoneda (1975) investigated the concentrations of heavy

metals in the sediments of rivers near Himeji City (west of Osaka); data from 30 samples revealed locally elevated concentrations of cadmium (0.56-10.4 ppm; 1.76-129 ppm at industrial plant sites), zinc (54-1840; 524-3240 ppm), copper (95-439 ppm; 22-776 ppm), and lead (17-607 ppm; 50-1888 ppm in industrial areas). Another crucial point of the investigations had to do with the rivers running through the mountainous regions of Central Japan, emptying into the Bay of Toyama. It was in this district that the catastrophic Cd-poisoning first appeared, leading to the sickness known as Itai-Itai. In a study on the sediments of the Takahara River, which becomes the Jintsu River upon confluence with the Miya River, Kiba et al. (1975) found near the Kamioka mine values of cd to be 43 ppm, Zn 6100 ppm, Pb 1500 ppm, Cu 740 ppm. Still higher values of lead, zinc and cadmium were recorded in the river sediments, their source being the abandoned mines of Hiraya on the upper course of the Takahara River and of Hirayu on the upper course of the Kohachiya River. Samples from 91 rivers in the Toyama Prefecture were studied by Shibahara et al. (1975) indicating heavy pollution with organic matter and metals in the Rivers Tsurumi, Yodo and Jintsu. Metal pollution from mercury bearing industrial effluents was evidenced by Matida and Kumada (1969) in the sediment of the River Agano (draining the Bay of Niigata) and by Sasaki et al. (1975) in the River Muka from a mercury-mining area of Hokkaido; in the latter deposits Hg occurs as a dispersion of annabar with grain sizes of 5-10 mm.

In Taiwan, rivers flowing into Kaohsiung Harbor (e.g., Ch'ien Chen River), contain elevated metal concentrations, partly traceable to both domestic sewage and industrial waste (Hung et al., 1977). High Hg concentrations were also recorded in the Philippines in river sediments close to chemical and plastic industries and paper and pulp plants (Lesaca, 1977). Sediment samples from the Balagtas River, where an industrial plant of International Chemical Industries is situated, revealed mercury concentrations of up to 642 ppm.

Trace metal analyses of sediments have been utilized to evaluate the distribution of sewage sludge in an Indonesian

stream (Houtman, 1973). In Thailand, heavy metal studies were undertaken to assess the environmental consequences of a proposed hydroelectric scheme in the River Quae Yai, thereby meeting the requirements of the World Bank with regard to financial assistance (McGarry et al., 1975).

The deleterious effects on ecosystems have been reported from the disposal of mining wastes in Bougainvillea, Papua, New Guinea (Brown, 1974). Accumulation of heavy metals in river sediments, originating from current mining activities, were found in Australia and Tasmania (Weatherley and Dawson, 1973; Tyler and Buckney, 1973) and in South Africa (Wittmann and Forstner, 1976, 1977).

Droppo and Ongley (1992) studied the significance of flocculated sediment in the fluvial environment of Oakville Sixteen Mile Creek in Ontario using digitizing method which allowed direct observation of floc structure which is impossible with conventional methods of sediment sizing. The method provides reasonable results with good precision on floc equivalent spherical diameter, surface area, perimeter and circularity.

Recently Bertin and Bourg (1995) investigated the trends in the heavy metal content in the upper Lot river basin in southwestern France with a view to observe the evolution of the quality of sediments of the river which has been contaminated with heavy metals for several decades. The Riou Mort is fed by an aquifer recharged by water percolating through tailings of former smelting activities. Its water and especially its sediments are rich in heavy metals (Say, 1981; Boisseau, 1987; Lotouche et al., 1989; Simonet, 1991; Bertin et al., 1992). The high metal concentrations in the sediments is reported to be associated with iron, manganese, organic and inorganic carbon, calcium, sulfide and small particles.

The significance of suspended and bed sediments in aquatic environment has long been recognized. They are also important vectors for the transport of contaminants in river systems. Pollutant concentrations in particulate matter often provide a more stable and convenient means of obtaining an indication of the state of associated waters. A number of studies on river and lake sediments have been conducted under both scientific aspects and direct pollution control which define the importance of metal pollution assessment through aquatic sediments. The analysis of sediments eliminate the problem of erratic fluctuations which are often observed in water and can be conveniently used to investigate possible pollution sources. The sediment existing at the bottom of the water body play a major role in the pollution schemes of the river and lake systems. The study of sediment cores provides a means to determine development of pollution intensities.

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